

#12



ATTORNEY DOCKET NO: 70181
IN THE UNITED STATES PATENT AND
TRADEMARK OFFICE

APPLICANT: Akihiro TADA and Tohru TSURUHARA

SERIAL NO.: 09/871,539

FILED: May 31, 2001

FOR: Monoazo metal complex compound containing composition and production method thereof

ART UNIT: 1756

EXAMINER: Janis L. Dote

Hon. Commissioner of Patents
and Trademarks

Washington, D.C. 20231

DECLARATION UNDER 37 CFR 1.132

I, Akihiro TADA, hereby declare as follows:

1. I graduated from Nagasaki University, Faculty of engineering, Department of Industrial Chemistry in March 1981. Since April 1981 I have been employed by Orient Chemical Industries Ltd. one of the assignees of the above-identified application where I have been engaged in research relating to charge control agent for toners.

2. I am one of the named inventors of the above-identified application and am familiar with the subject matter disclosed in said application as well as the disclosures in the references cited against the claims including the patent to Ookubo et al., USP No. 6,120,958 and the patent to Sukata et al., USP No. 5,843,611.

3. In order to demonstrate the difference between the invention of the above-identified application and the cited Ookubo 958 patent and Sukata patent, the following experiments were conducted:

Experiments

Synthesis of monoazo dye

Water 600 ml

Hydrochloric acid 40.0 g

4-chloro-2-aminophenol 57.6 g (0.4mol)

After an aqueous solution comprising the above ingredients was cooled to 5°C, a solution of 28.0 g (0.405mol) of sodium nitrite in 90 ml of water was added to the solution drop by drop over a period of 30 minutes. This mixture was stirred at 5 to 15°C for 1 hour, after which the reaction mixture was filtered, to yield an aqueous solution of the diazonium salt of 4-chloro-2-aminophenol.

Water 800 ml

Sodium hydroxide 28.0 g

2-naphthol 57.6 g

Subsequently, the aforementioned solution was added drop by drop over a period of 40 minutes to an aqueous solution of the ingredients shown above, after which this mixture was stirred for 3 hours. The precipitated reaction product was collected by filtration, washed with water, and dried at 100°C, to yield 126.5 g of 1-(5-chloro-2-hydroxyphenyl)azo-2-hydroxynaphthalene (monoazo dye).

Synthesis of monoazo metal complex compound

Monoazo dye obtained above 126.5 g

Chromium formate 38.4 g

Ethylene glycol monoethyl ether 400 g

After the above ingredients were mixed and stirred at 130°C for 6 hours, this mixture was cooled to 90°C, and the reaction mixture was dispersed in an aqueous solution of 30 g of hydrochloric acid in 400 ml of water. The dispersion liquid was filtered to obtain a wet cake, which was divided into quarters, i.e. wet cakes A, B, C and D. Each wet cake was subjected to the undermentioned washing process respectively.

(A) Composition A

Wet cake A was dried at 100°C to obtain a black product, which was milled to yield 31.0 g of monoazo metal complex compound containing composition A.

(B) Composition B

Wet cake B was dried at 100°C to obtain a black product, which was milled to a black powder. 31.0 g of the black powder was dispersed in 80 g of methanol and stirred at 60°C for 1 hour, after which it was cooled to 30°C and filtered through Toyo Filter Paper No. 2. The wet cake obtained was further washed with 20 g of methanol on filter paper and dried at 50°C under reduced pressure to yield 28.1 g of monoazo metal complex compound containing composition B.

(C) Composition C

Wet cake C was dispersed in 750 ml of water and stirred at 60°C for 1

hour, after which it was filtered. The wet cake obtained was further washed with 50 ml of water and dried at 100°C to obtain a black product, which was milled to yield 30.2 g of monoazo metal complex compound containing composition C.

(D) Composition D

Wet cake D was dispersed in a mixture of 200 ml of water and 40 g of ethanol and stirred at room temperature for 1 hour, after which it was filtered. The wet cake obtained was washed with 50 ml of water and dried at 100°C to obtain a black product, which was milled to yield 29.5 g of monoazo metal complex compound containing composition D.

Each of Compositions A, B, C and D was analyzed by high performance liquid chromatography and purity of monoazo metal complex compound in each composition was determined as presented in the table 1.

High performance liquid chromatography (HPLC) operating conditions

Column: L-column ODS (produced by the Chemicals Inspection & Testing Institute, Japan)

Eluent: Mixture of 80 parts by volume of acetonitrile, 1 part by volume of triethylamine, 3 parts by volume of acetic acid, and 16 parts by volume of water

Flow rate: 1.0 ml/min

Column temperature: 45°C

Measuring wavelength: 313 nm

Injection volume: 10 μ l

Sample preparation: 10 mg/10 cc (80 parts by volume of acetonitrile and 20

parts by volume of water)

Table 1

	HPLC determined purity of monoazo metal complex compound (%)
Composition A	81.3
Composition B	95.1
Composition C	83.4
Composition D	87.8

4. I hereby declare that all statements made herein of my own knowledge are true and that all statements made on information and belief are believed to be true; and further that these statements were made with the knowledge that willful false statements and the like so made are punishable by fine or imprisonment or both under Section 1001 of Title 18 of the United States Code and that such willful false statements may jeopardize the validity of the application or any patent issued thereon.


Akihiro TADA

Chief Researcher of Developing Charge Control Agent for Toners

Laboratory of The Fourth Technical Development Department

Dated: Apr 2 , 2003